EFFECT OF MAGNETIC FIELD ON INDIRECT EXCHANGE INTERACTION OF LOCALIZED SPINS IN A METAL

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An expression for indirect exchange interaction between spins is obtained by taking into account magnetic quantization in metals and semiconductors. Besides the oscillations of exchange-interaction accompanied by variation of the magnetic field in metals, which are analogous to the De Haas—van Alphen effect oscillations, the magnetic field leads to the appearance of the following features of the interaction: 1) the interaction between a pair of spins located in a plane perpendicular to the magnetic field, $\mathbf{R}_{ji} \perp \mathbf{H}$, is ferromagnetic at arbitrary values of $R \equiv |\mathbf{R}_{ji}|$, whereas the sign of the interaction in a zero field depends on R; 2) the magnitude of the interaction in a field with $\mathbf{R}_{ji} \parallel \mathbf{H}$ decreases with increasing R like R^{-1} or R^{-2} but not like R^{-3} in the absence of the field.

INDIRECT exchange interaction between nuclear spins via the conduction electrons was considered in[1-4]. A similar interaction between paramagnetic impurities in a nonmagnetic metal, as a result of s–f(d) exchange interaction of the impurity ions with the conduction electrons, was investigated in[5-6].

In the theory of the line shape of nuclear magnetic resonance in heavy metals, the anomalously large widths of the resonance lines are attributed to the existence of indirect interaction between the nuclei. Indirect exchange interaction between electrons of the f(d) shells plays an important role in the interpretation of the magnetic properties of metals of the rare-earth group[7], and also of the ferromagnetic properties of alloys.

In second order of perturbation theory, the operator of interaction between the spins localized in the sites j and i is given by the expression

$$H_{ji}=\sum_{\substack{k,\beta\\k',\beta'}}\frac{\langle k,\beta|V(j)|k',\beta'\rangle\langle k',\beta'|V(i)|k,\beta\rangle}{\varepsilon_\beta(k)-\varepsilon_{\beta'}(k')},\qquad(1)$$

where V(j) is the operator of spin interaction at the site j with the conduction electrons, k and k′ are the momenta of the electrons with spin β and β', and $\epsilon_\beta(k)$ is the energy of the electrons in the state k, β. In all the foregoing studies of the exchange interaction, the usual assumptions were made concerning the sphericity of the Fermi surface, and the conduction electrons were described by a plane Bloch function. In this case the interaction takes the form

$$H_{ji}=-\frac{m}{8\pi^2\hbar^2}A^2(\mathbf{J}_j\mathbf{s}_{\beta\beta'})(\mathbf{J}_i\mathbf{s}_{\beta'\beta})\frac{\sin k_FR-2k_FR\cos 2k_FR}{R^4},\qquad(2)$$

where A is the constant of interaction between the conduction electrons and the localized spin, m is the electron effective mass, J are the angular momentum operators of the localized spins, $s_{\beta\beta'}$ are the matrix elements of the spin operators of the conduction electrons, and $R = |\mathbf{R}_{ji}|$.

The expression for H_{ij} depends on the energy spectrum and on the character of the wave functions describing the conduction electrons. As is well known[8], the energy spectrum and the wave functions of the conduction electrons change greatly when a metal or alloy is placed in the magnetic field. We consider in this paper the influence of a quantizing magnetic field on the indirect exchange interaction between localized magnetic moments in metals and semiconductors. For an isotropic conductor with quadratic dispersion law in a magnetic field A = (0, xH, 0) the normalized wave functions and the electron spectrum are given by

$$\psi_{n,k_z}=\Omega_n^{-1/2}\exp(ik_xx+ik_zz)\exp[-{}^1/_2\alpha(y-y_0)^2]H_n(\sqrt{\alpha}(y-y_0)^2)u(r),$$

$$\varepsilon_{n,k_z}=\left(n+\frac{1}{2}\right)\hbar\omega_0+\frac{\hbar^2}{2m}k_z^2-g\beta Hs_z^\beta,\qquad(3)$$

where $\Omega_n = 2^n n!\sqrt{\pi}L_xL_y\alpha^{-1/2}$, $y_0 = -k_x/\alpha$, $\alpha = eH/\hbar c$, $H_n(x)$ is the Hermite polynomial, n is the number of the Landau level, $\omega_0 = eH/mc$, and L_x, L_y, L_z are the linear dimensions of the sample.

The operator of interaction between the localized magnetic moment and the electrons is written in the form

$$V=-\frac{I}{N}\sum_j(\mathbf{J}_j\mathbf{s})\delta(\mathbf{r}-\mathbf{R}_j),\qquad(4)$$

where I is the exchange-interaction integral.

In the general case allowance for the anisotropy of the distribution of the electrons in the f (or d) shell, and for the component with the orbital angular momentum $l \neq 0$ in the wave function of the conduction electron, leads to a complicated dependence of I(k, k′) on the vectors k and k′:

$$I(\mathbf{k},\mathbf{k}')=\int\psi_d^*(\mathbf{r}_1)\varphi_{\mathbf{k}'}^*(\mathbf{r}_2)\frac{e^2}{r_{12}}\psi_d(\mathbf{r}_2)\varphi_{\mathbf{k}}(\mathbf{r}_1)d\mathbf{r}_1\,d\mathbf{r}_2.$$

However, as shown in[9], under the condition $\overline{r_j^2}k_F^2 < 1$, where $\overline{r_f^2}$ is the mean square of the radius of the f(d) shell, it is sufficient to confine oneself to the first term of the excitation of I(k, k′) in spherical harmonics:

$$I(\mathbf{k},\mathbf{k}')=\sum_n I_nP_n(\cos\theta),$$

where θ is the angle between the vectors k and k′ (for gadolinium, for example, $I_1/I_2 = 0.2$[10]).

If we consider the interaction between the nuclear spins of the conduction electrons, the exchange integral in (4) is given by

$$I = -{}^{8}/{}_{3}\pi\mu_N\mu_e,$$

where μ_e and μ_N are the magnetic moments of the electron and of the nucleus, respectively. Substituting (4) and (3) in the expressions for $H_{ji}(1)$ and integrating with respect to k_x, k'_x and k_z, k'_z for the case when $\epsilon_F \gg h\omega_0$ and $kT < h\omega_0$, we obtain

$$H_{ji} = \frac{m}{2\pi^3\hbar^2}\left(\frac{I}{N}\right)^2 (\mathbf{J}_j\mathbf{s}_{\beta\beta'})(\mathbf{J}_i\mathbf{s}_{\beta'\beta})F(x, z, \alpha),$$

$$F(x, z, \alpha) = \alpha^2 \sum_{n_1, n_2} l_{n_1}\left(\frac{x^2\alpha}{2}\right) l_{n_2}\left(\frac{x^2\alpha}{2}\right) \mathrm{si}\,[z(k_z(n_1) + k_z(n_2))], \tag{5}$$

where $l_n(x)$ is the Laguerre function, $l_n(x) = e^{-x/2}L_n(x)$, $N_n(x)$ is the Laguerre polynomial, $x = |X_{ij}|$, $z = |Z_{ij}|$, $0 \le n_{1,2} \le N_{\beta\beta'}$,

$$N_\beta = \mathrm{E}\left(\frac{\varepsilon_F + \Delta_\beta}{\hbar\omega_0} - \frac{1}{2}\right), \quad \frac{\Delta_\beta}{\hbar\omega_0} = g s_z{}^\beta \frac{m}{m_0},$$

$$k_z(n) = \left\{\frac{2m}{\hbar^2}\left[\varepsilon_F - \left(n + \frac{1}{2}\right)\hbar\omega_0 + \Delta_\beta\right]\right\}^{1/2},$$

m_0 is the mass of the free electron, and E denotes the integer part. Summation over β and β' is implied in (5). To simplify the notation, we have assumed here $y_i = y_j = 0$. This does not limit the generality, in view of the invariance of the Hamiltonian against rotations around the z axis (H ∥ z), $x^2 \to x^2 + y^2$. In integrating with respect to k_x and k'_x in (1), it was assumed that $L_y - y_0 \gtrsim \alpha^{-1/2}$ (for $H = 10^4$ G, we have $\alpha^{-1/2} \approx 10^{-5}$ cm), i.e., the radii of the quasiclassical orbits of the electrons in the magnetic field do not touch the surface of the sample. By the same token, we exclude from consideration effects due to surface magnetic levels[11-13]. In the integral

$$B_n = l_n\left(\frac{x^2\alpha}{2}\right) = \frac{1}{\alpha}\int_{-k_0}^{k_0} dk_x \cos k_x x \exp\{-\alpha y_0^2\}\,[H_n(\sqrt{\alpha}\,y_0)]^2,$$

where $k_0 = \alpha L_y/2$, the integration limits can be regarded as infinite.

Let us consider separately two cases: 1) $m = m_0$, $\epsilon_F/\hbar\omega_0 \approx 10^4$—$10^6$; 2) $m = 10^{-2}m_0$, $\epsilon_F/\hbar\omega_0 \lesssim 10$. In most metals, the conditions indicated for the first case are satisfied.

1. In an analysis of the dependence of H_{ji} and the magnetic field and of the distance R_{ji} between the spins for the case $N_\beta \gg 1$, it is convenient to set one of the variables, z or x, equal to zero. When z = 0 we have $\mathrm{Si}(z) = -\pi/2$, and the sum over n_1 and n_2 can be readily calculated[14]:

$$\sum l_{n_1}(x)\, l_{n_2}(x) = l_{N_1}{}^1(x)\, l_{N_2}{}^1(x).$$

Using the asymptotic expression for the Laguerre polynomials at large N:

$$L_N{}^1(t) = \pi^{-1/2}e^t t^{-3/4}N^{1/4}\cos(2\sqrt{tN} - 3\pi/4) + O(N^{-1/4}), \quad t = \alpha x^2/2,$$

and bearing in mind the fact that $N_{\beta'} = N_\beta \pm (1 - \delta_{\beta\beta'})$ when $m = m_0$ and $s_z = \pm 1/2$, we obtain

$$H_{ji} = -\frac{m}{4\pi^3\hbar^2}\left(\frac{I}{N}\right)^2 (\mathbf{J}_j\mathbf{s}_{\beta\beta'})(\mathbf{J}_i\mathbf{s}_{\beta'\beta})F(k_F, x, \alpha), \tag{6}$$

where

$$F(k_F, x, \alpha) = \frac{k_F}{x^3}\{\sin[x(2\alpha)^{1/2}(\sqrt{N_\beta} + \sqrt{N_{\beta'}})] + \cos[x(2\alpha)^{1/2}(\sqrt{N_\beta} - \sqrt{N_{\beta'}})]\}.$$

As seen from (6), the dependence of the indirect exchange interaction on R, for impurities in a plane perpendicular to the magnetic field, coincides qualitatively with the dependence of H_{ji} when H = 0. However, the value of H_{ji} oscillates when the magnetic field is varied. When g = −2, the amplitude and period of the oscillations are determined by the factor $\sin(2x\sqrt{2\alpha N_\beta})$. The period of the H_{ji} oscillations is determined from the relation $N_\beta(H_0) = N_\beta(H_0 + \Delta) - 1$ (where H_0 is the value of the field at which $N_\beta = \epsilon_F/\hbar\omega_0 - 1/2$), and is equal to $\Delta = eH_0^2(c\hbar k_F^2)^{-1}$. For K, Cu and Ag, with Fermi energies equal to 5.5, 7, and 2.4 eV respectively, the period Δ at $H = 2 \times 10^4$ G takes on the values 0.5, 0.17, and 0.25 G. The amplitude of the oscillations is determined by the difference

$$\sin 2xk_F - \sin 2x\sqrt{2\alpha(H_0 + \varepsilon)N(H_0)}, \quad 0 \leqslant \varepsilon \leqslant \Delta$$

and reaches its maximum value at $x(2\alpha)^{1/2} = \pi k_F/2$. For real values of the constants in (6) ($k_F = 10^8$ cm^{-1}, impurity concentration $c \gtrsim 0.1$ at.%, $x \approx 10^{-6}$ cm, I = 0.5 eV, $H_{ji} \approx 10^{-17}$ erg), the oscillation amplitude is such that $\Delta H_{ji}/H_{ji} \approx 10^{-2}$—$10^{-3}$.

The summation over n_1 and n_2 in the case when x = 0 and z ≠ 0 can be replaced by integration, by first eliminating from the sum the term with $n = N_{\beta,\beta'}$. Retaining the principal terms in $F(k_F, z, \alpha)$, we obtain

$$-\frac{1}{6}\frac{k_F{}^2}{z^2}\left(\sin 2k_F z + \frac{\cos 2k_F z}{zk_F}\right). \tag{7}$$

The contribution made to the expression for $F(k_F, z)$ by terms oscillating with H is smaller by a factor 10^2—10^4 than the principal contribution.

It follows from relations (7) and (6) that when a conductor having a quadratic dispersion law is placed in a magnetic field the indirect exchange interaction acquires the following features: 1) the magnitude of the interaction oscillates with the magnetic field as expected, in general, from the analogy with de Haas—van Alphen effect; 2) the interaction between the localized spins in a plane perpendicular to the magnetic field is ferromagnetic, whereas at H = 0 the sign of the interaction depends on the quantity $k_F R$; 3) when $\mathbf{R}_{ji} \parallel \mathbf{H}$ and $N_\beta \gg 1$ the interaction of the pair of spins decreases with the distance between them like R^{-2}, and not like R^{-3} in the absence of a field.

The results can be interpreted in the following manner: The indirect exchange interaction, due to the exchange of virtual electrons pertaining to the Landau levels n_1 and n_2, is proportional to the electron density matrix of the j-th and i-th sites $\rho(i, j) = \psi^*(i)\psi(j)$. Since the wave function of the electrons rotating in a magnetic field on a quasiclassical orbit with center y_0 differs from zero in a radius $r \approx (2\alpha)^{-1/2}(2n + 1)^{1/2}$ in the vicinity of y_0, it is obvious that the main contribution to the interaction is made by electrons with $r \le X_{ji}$. The wave function of the electrons located at the Landau levels with $n \approx N_\beta \gg 1$ oscillates with varying distance from the center y_0 like $\cos[(y - y_0)\sqrt{4n + 1}]$. When the magnetic field changes, $H_0 \to H_0 + \Delta$, the strongest change occurs in the distribution of the electrons located at the levels $n \approx N_\beta$. The relative contribution of such electrons located at the levels $n \approx N_\beta$. The relative contribution of such electrons to the indirect interaction increases with increasing x, and therefore when $x \gtrsim r_{max} \approx \alpha^{-1/2}N_\beta^{1/2}$ the amplitude of the variation of H_{ji} with changing magnetic field reaches its maximum value.

The contributions of the oscillating density matrices $\rho(i, j) \approx \cos kr$, pertaining to two different states participating in the virtual exchange, are not independent when H = 0, and are connected in (1) by the energy factor $\epsilon(k) - \epsilon(k') < 0$, which takes on only negative values when $kT < \hbar\omega_0 \ll \epsilon_F$ (transitions are possible only to the upper unoccupied levels). The interference of the contributions of the states with different k and k′ leads to oscillations of the sign and magnitude of H_{ji}. When $2k_FR < \pi$, for example, $\cos 2k_FR$ assumes only positive values, and therefore the sign of the interaction is determined by the sign of the energy factor and, as can be readily verified, the interaction (2) is ferromagnetic at small values of k_FR. Magnetic quantization causes the energy factor to become independent of n_1 and n_2 when H = 0; the interaction is in this case proportional to

$$\Big(\sum_{n,\,n'} \rho_{nn'}(i,j)\Big)^2 = \Big(\sum_n B_n\Big)^2,$$

and its sign is determined by the energy factor.

The appearance in the expression for H_{ji} of terms that decrease with increasing distance more slowly than R^{-3} is a general rule when the quadratic dispersion function ceases to be a smooth function of the particle momentum[15]. In a magnetic field, the values of the z component of the electron wave vector in the interval $|k_z| \le k_F$ may not constitute a continuous series, and the continuous interval, depending on the number of the Landau level, breaks up into a series of discrete intervals

$$|k_z(n)| \leqslant (2m/\hbar^2)^{1/2}[\varepsilon_F - (n + 1/2)\hbar\omega_0]^{1/2}.$$

The limiting transition $H \to 0$ in relations (6) and (7) is incorrect and does not lead to expression (2) for H_{ji} (H = 0), since at low magnetic field intensities, $H \le k_F c\hbar e^{-1}$, the radii of the helical trajectories become comparable with the dimensions of the sample and the wave functions (3) are not suitable in this case for the description of the electronic states.

It is impossible to sum the series (5) with sufficient accuracy in the case when x, z ≠ 0. A rough estimate of F(x, z) can be obtained for n > 1 after carrying out the calculations with the aid of the Abel transformation[16]

$$\sum_n f(n)\varphi(n) = F(N)\varphi(N) - \int_1^N F(x)\varphi'(x)\,dx, \quad F(x) = \sum_1^x f(n), \quad (7')$$

which leads to cumbersome expressions that will not be written here. It is of interest, however, to estimate in which intervals of angles to the chosen directions along and across the field one can observe the aforementioned changes in the character of the interaction. Expanding the integral sine function in expression (5) in a series in small values of $z = R\tan\theta$ (R—distance between impurities) and summing with the aid of the Abel relation (7′) a series in the form

$$\sum l_n(y)\,l_{n'}(y)\sqrt{N-n},$$

we find that the ferromagnetic character of the exchange interaction is retained near the direction $\mathbf{R} \perp \mathbf{H}$ in the angle interval

$$|\theta| \leqslant \operatorname{arc\,tg}\Big(\frac{\pi}{6\sqrt{2}}\frac{1}{Rk_F}\Big).$$

Carrying out similar calculations for small values of x, we find that the R^{-2} dependence for the ordinary interaction remains in force when **R** deviates from the direction of H in the interval $|\varphi| \le \tan^{-1}(\sqrt{5/2}/Rk_F)$.

In metals with $m = m_0$ and $e_F/\hbar\omega_0 \gg 1$, as is well known, it is difficult to realize the conditions under which the motion of the electrons in a magnetic field is quantized

$$\hbar\omega_0 > kT, \quad \omega_0 > \tau^{-1}$$

(τ—relaxation time); therefore, from the experimental point of view, great interest attaches to metals and semiconductors with $m \ll m_0$ and $\epsilon_F/\hbar\omega_0 \approx 10$.

2. In metals, semimetals, and degenerate semiconductors with $m/m_0 \ll 1$, it may turn out that the dispersion law is quadratic but anisotropic:

$$\varepsilon = \frac{p_1^2}{2m_1} + \frac{p_2^2}{2m_2} + \frac{p_3^2}{2m_3}. \qquad (8)$$

This condition is satisfied for Bi, Sb, and As if we neglect the deviation of one of the axes of the effective-mass ellipsoid from the trigonal axis by 6°[17]. The condition (8) can also be extended to Ga, Zn, and C (graphite). Expression (5) remains in force for such samples, but in this case we have in (5)

$$\frac{2m}{\hbar^2}a^2 \to \frac{2}{\hbar^2}\frac{m_1m_2m_3}{m^{*2}}\Big(\frac{eH}{c\hbar}\Big)^2, \qquad z \to \sum_i \Big(\frac{m_i}{m}\Big)^{1/2}\alpha_i x_i,$$

$$\alpha x^2 \to \Big(\frac{eH}{c\hbar}\Big)^2\frac{1}{m^*}\Big[\sum_i (1-\alpha_i^2)m_i x_i^2 - 2\sum_{i,j}\alpha_i\alpha_j(m_im_j)^{1/2}x_ix_j\Big],$$

$$m^{*-1} = \Big(\frac{\alpha_1^2}{2m_2m_3} + \frac{\alpha_2^2}{m_1m_3} + \frac{\alpha_3^2}{m_1m_2}\Big)^{1/2}, \qquad (9)$$

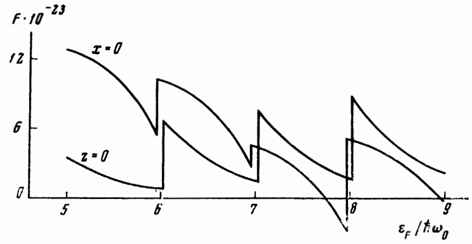
where α_i are the direction cosines of the magnetic field relative to the principal axes of the effective-mass ellipsoid. The indirect exchange interaction in such substances is anisotropic and depends on the direction of the magnetic field, $N_\beta = N_\beta(H)$.

The EPR line width of impurities in metals is due to the following processes[18,19]: spin relaxation as a result of the s-d exchange interaction, magnetic dipole-dipole interaction, and indirect exchange interaction. According to the measurements[18,19] carried out at temperatures T ≤ 5–10°K, the EPR line width is determined by the pair interactions of the impurities. When $N_\beta \le 20$, the asymptotic expressions (6) and (7) are not suitable.

The table lists the values of the constants in expressions (5) and (2) for semimetals that are apparently the most suitable for an experimental observation of the dependence of H_{ji} on H. The values of the constants were taken from[20]. For samples with impurity concentration c ≈ 2–0.1 at.% (R ≈ 5×10^{-6} cm), the energy of the

	m_1/m_0	m_2/m_0	m_3/m_0	E_F, 10^{-14} erg
Al	$8\cdot10^{-3}$			6
Ga	0,2	0.02	0.4	4.6
Zn	$5.3\cdot10^{-3}$	1.0	0.2	4.9
Bi	$2.4\cdot10^{-3}$	2.5	0.05	2.9

indirect exchange interaction is equal to $H_{ji} \approx 10^{-22}-10^{-23}$ erg, which is larger by one order of magnitude than the energy of the dipole-dipole interaction at the same distance, $H_d \approx 3 \times 10^{-24}$ erg.



We choose the direction of the magnetic field along the axis with $m_i = m_{max}$. The difference in the values of N_β and $N_{\beta'}$, which is equal to $m^*/m_0 \approx 10^{-2}$, can be neglected. Using the recurrence relations for the Laguerre polynomial[14],

$$l_N{}^1(y) = -\frac{N+1}{y}[l_{N+1}(y) - l_N(y)],$$

we obtain for the function F, which describes the dependence of H_{ji} on the field at z = 0,

$$F = \frac{(N+1)^2}{x^4}\left[l_{N+1}\left(\frac{x^2\alpha}{2}\right) - l_N\left(\frac{x^2\alpha}{2}\right)\right]^2.$$

The figure shows plots of $F(\alpha, x, z)$ against $\epsilon_F/\hbar\omega_0$ ($\omega_0 \approx H^{-1}$) at different orientations R_{ij} relative to H.

An expression for the indirect exchange interaction in nondegenerate semiconductors at H = 0 was obtained in[21]. Integration with respect to k_z and k_z' for a nondegenerate semiconductor leads to replacement of si z (n, n′) in (5) by

$$\exp(-\gamma B_1{}^2)C_1 + \exp[-\gamma(B_1{}^2 + B_2{}^2)/2] \operatorname{si} az,$$

where

$$B_{1,2}^2 = \frac{2m}{\hbar^2}\left[E_c + \left(n_{1,2} + \frac{1}{2}\right)\hbar\omega_0 + \Delta_{\beta,\beta'} - E_F(H)\right], \quad \gamma = \frac{\hbar^2}{2m}\frac{1}{kT}$$

$$a = \sqrt{B_1{}^2 - B_2{}^2}, \quad n_1 \geqslant n_2,$$

$$C_1 = \frac{1}{e^{-b} + \gamma b}\sqrt{\frac{\pi}{2}}\, e^{-z^2/\gamma}\left[\sin az + \frac{z}{\sqrt{\gamma}}\Phi\left(\frac{1+e^{-b}}{2}, \frac{3}{2}, \frac{z^2}{\gamma}\right)\right],$$

E_c is the energy of the electron at the bottom of the conduction band, $\Phi(\alpha; \gamma; \beta)$ the confluent hypergeometric function, and $b = \sqrt{a^2/\gamma}$.

Unlike metals, where all the magnetic levels up to the Fermi level are filled when $p = \hbar\omega_0/kT \gg 1$, only the lower levels are mainly populated in semiconductors at the same temperatures. The dependence of the indirect exchange interaction on the magnetic field in a nondegenerate semiconductor is determined mainly by statistical parameters—the change of the population of the magnetic levels and of the total number of electrons in the conduction band with changing field.

The oscillatory effects due to the change of the wave function of the electrons participating in the exchange, at $R \approx 10^{-5}-10^{-6}$ cm, have large periods, $\Delta \approx 5 \times 10^3$ G, when p > 1, and are determined by the relation

$$[a(H+\Delta) - a(H)]z \approx \pi/2.$$

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